PATENT SPECIFICATION (11) 1361402

(19)

(54) IMPROVEMENTS IN OR RELATING TO THE HEAT TREATMENT OF PARTICULATE MATERIALS

ERRATUM

SPECIFICATION No. 1,361,402

Page 3, line 79, *after* was *insert* 1100°C. Air
flow through the fluidised bed was

THE PATENT OFFICE
23rd December, 1974

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10 of particulate materials and, more particularly   fluidised bed for a time sufficient to effect the
but not exclusively, is concerned with the cal-   desired heat treatment thereof.

PATENTS ACT 1949

SPECIFICATION NO 1361402

The following amendments were allowed under Section 29 on 16 June 1975

Page 1, line 36, page 8, line 3, *after* material *insert* which is a silicate of
aluminium or of an alkaline earth metal and

Page 2, *delete* lines 53, 54, 55 and 56, *insert* range of from 600° to 1200°C.
The average residence time of the particles of a particulate material within the

Page 8, *delete* lines 52, 53, 54 and 55

Page 8 *for* claims 9 to 20 *read* 8 to 19 inclusive

Page 8, line 56, *delete* Claim 8 *insert* any one of the preceeding claims

Page 8, line 67, *delete* 10 *insert* 9

Page 8, line 90, *delete* 16 *insert* 15

Page 8, line 103, *after* material *insert* which is a silicate of aluminium or of
an alkaline earth metal

Page 8, *delete* lines 106, 107, 108 and 109

THE PATENT OFFICE
July 1975

R 23917/6

~~ is passing and through a major proportion by   of the dense-phase type, but spouting fluidised
weight of said inert refractory material; (ii)   beds can also be used.

SEE ERRATA SLIP ATTACHED

SPECIFICATION AMENDED - SEE ATTACHED SLIP

PATENT SPECIFICATION (11) 1361402

(54) IMPROVEMENTS IN OR RELATING TO THE HEAT
TREATMENT OF PARTICULATE MATERIALS

(71) We, ENGLISH CLAYS LOVER-ING POCHIN & COMPANY LIMITED, a British company, of John Keay House, St. Austell, Cornwall, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to the heat treatment of particulate materials and, more particularly but not exclusively, is concerned with the calcining of minerals such as the silicates of aluminium and of the alkaline earth metals.

The heat treatment of minerals such as the silicates of aluminium and of the alkaline earth metals by an operation known as "shock calcination" is disclosed in British Patent Specifications Nos. 866,326 and 869,966 and essentially comprises the spraying of the silicates into a vortex of hot gases. These known processes, however, suffer from the disadvantage that the temperature varies within the reaction vessels and cannot be accurately controlled. Furthermore, the residence time of particles in the reaction vessels varies over a wide range and there is a strong tendency for the particles to agglomerate so that the particle size of the product also varies over a wide range.

It is an object of the present invention to provide a process for the heat treatment of particulate materials in which the above disadvantages can be overcome or inexacerbated.

According to the present invention there is provided a process for heat treating, e.g. calcining, a particulate material which consists predominantly of particles smaller than 50 microns equivalent spherical diameter, which process comprises passing the particulate material through a heated fluidised bed comprising particles of an inert refractory material which range in diameter from 0.5 to 5.0 mm, wherein (i) the fluidised bed contains throughout the period in which the particulate material is passing therethrough a major proportion by weight of said inert refractory material; (ii)

the size, shape and density of the particles of inert refractory material and the velocity of the fluidising gas are such that the particles of inert refractory material and the velocity 50 of the fluidising gas are such that the particles of inert refractory material are retained in the fluidised bed while the particulate material is carried through the fluidised bed; and (iii) the particulate material is retained within the 55 fluidised bed for a time sufficient to effect the desired heat treatment thereof.

Preferably, the inert refractory material consists of particles having a definite but narrow particle size distribution range, it being advantageous for the range of particle size of 60 the inert refractory material to be such that the coarsest particles of the inert refractory material are not more than four times larger than the finest particles of the inert refractory 65 material. It is also preferable for the shape and average particle size of the inert refractory material to be such that the linear gas velocity necessary to fluidise the inert refractory material is at least five times that neces- 70 sary to convey the particulate material out of the fluidised bed.

The inert refractory material can be, for example, sand, silica or a ceramic material. Good results have been obtained when using 75 as the inert refractory material a calcined kaolin clay, for example that sold under the Trade Mark "MOLOCHITE".

Advantageously, the particles of the particulate material to be heat treated are fed 80 pneumatically into the fluidised bed; but other feed methods, for example gravity feed, can alternatively be employed. It has also been found to be advantageous to feed the particles of the particulate material to be heat treated 85 via a conduit passing longitudinally through the top of the fluidised bed reactor and extending into the fluidised bed, although laterally positioned conduits have also functioned satisfactorily. The fluidised bed is advantageously 90 of the dense-phase type, but spouting fluidised beds can also be used.

SEE ERRATA SLIP ATTACHED

A fluidised bed is formed when a fluid flows upwardly through a bed of suitably sized solid particles at a velocity sufficiently high to buoy the particles, to overcome the influence of gravity thereon, and to impart thereto an appearance of great turbulence.

The fluidising gas used to form the fluidised bed can be air, and advantageously the fluidising gas is also used to support the combustion of a fuel which provides the heat for the fluidised bed. The heating of the fluidised bed can be achieved, for example, by using hot combustion gases as the fluidising gas or by burning a fuel within the fluidised bed itself. This latter method is preferred because it allows the use of a fluidisation grid, or distribution plate, which does not need to withstand very high temperatures, as well as giving a more even temperature distribution within the fluidised bed. This allows a more evenly heat treated, e.g. calcined, product to be obtained. More preferably, the fluidised bed is initially heated by passing therethrough the combustion gases obtained from a conventional combustion chamber burning a liquid, gaseous or powdered solid fuel, and, when the temperature of the fluidised bed approaches the desired working temperature, the supply of combustion gases is stopped and fuel is injected into the fluidised bed, or at a point just below the fluidisation grid, so that ignition and combustion of the fuel takes place in the fluidised bed itself. The fuel injected into the fluidised bed can be liquid, gaseous or a powdered solid. One or more further fluidised beds can be provided above the first fluidised bed to ensure complete combustion of all the fuel injected into the lower bed.

Preferably, the particles of the particulate material which are fed into the fluidised bed reactor should be dry or substantially dry. However, in certain circumstances, for example when the particulate material that is to be heat treated is to be used to form a cement, a slurry of the particulate material can be fed into the fluidised bed. This latter mode of operation, however, requires a high degree of heating if high temperatures are to be maintained within the fluidised bed.

Generally, if the particulate material to be heat treated is to be calcined, the fluidised bed will be operated at a temperature in the range of from 600° to 1200° C. When calcining silicates of the alkaline earth metals or of aluminium, for example, the average residence time of the silicate particles within the heated fluidised bed should generally be one second or less. The residence times of individual particles of a particulate material within the fluidised bed should not normally exceed three seconds. The residence time of the particulate material in the fluidised bed is governed by the bed depth and the velocity of the gases passing up through the bed; the smaller the bed depth and the greater the gas velocity, the shorter will be the residence time of the particulate material in the fluidised bed. The gas velocity is affected in turn by the size, specific gravity and shape of the particles of inert refractory material, since there is a minimum gas velocity required to fluidise the particles. For particles of a given specific gravity and shape, the fluidising velocity is less for small particles than for large particles.

Separation of the heat treated particulate material from the fluidising gas can be performed by known methods, for example by using a cyclone or two or more cyclones arranged in series or in parallel. Other types of separator, for example electrostatic precipitators or bag filters, may also be used either alone or in conjunction with a cyclone or cyclones. The suspension of heat treated particulate materials in the fluidising gas is preferably cooled to below 300° C. either before or after passing through the separator(s) by injecting a cold liquid, e.g. water or air, into the suspension. It is advantageous to provide an exhaust fan which will draw the suspension through the separator and maintain the pressure in the reaction vessel very slightly below atmospheric pressure so that the particulate material and inert refractory material are not blown out through the feed conduit.

The process of the present invention enables close control of the temperature at which the particulate material is treated to be obtained and allows efficient heat transfer between the particulate material and the inert refractory material. In addition, the turbulence within the fluidised bed tends to minimise the tendency of the particulate material being heat treated to agglomerate.

For a better understanding of the invention, and to show how the same can be carried into effect, reference will now be made, by way of example, to the accompanying drawings, which show four embodiments of an apparatus for use in the process of the present invention. Similar reference numerals in the Figures refers to similar parts of the apparatus.

Referring first to Figure 1, a fluidised bed of particles of inert refractory material 1 is supported on a perforated distribution plate 2 and is contained in a vessel 3 having thermally insulated walls 4. The fluidised bed is initially heated to near the working temperature by means of hot combustion gases from a combustion chamber 5 in which a fuel oil is burned in air. When the desired fluidised bed temperature has been reached, the supply of hot combustion gases is stopped and fuel oil is injected through a conduit 6 below the distribution plate of the fluidised bed, or through a conduit 7 above the distribution plate, or both. The particulate material which is to be heat treated, for example an uncal-

cined silicate, is injected as a suspension in air into the fluidised bed through a conduit 8. Heat treated, e.g. calcined, particulate material leaves the apparatus through a conduit 9 which communicates with a cyclone separator 10. The gases leave the cyclone through a conduit 11 and the heat treated particulate material collects in the bottom of the cyclone whence it can be removed by opening, in turn, valves 12 and 13. A sight glass 14 is provided in the top of the vessel 3 to permit visual inspection of the fluidised bed, and a discharge conduit 15 permits discharge of the particles of inert refractory material from the vessel.

Referring next to Figure 2, there is shown an embodiment of the invention in which the particulate material to be heat treated is injected into the fluidised bed, either through a conduit 18, which dips below the surface of the fluidised bed 1, or through a lateral conduit 8. This embodiment has a single fuel conduit 7.

Referring next to Figure 3, there is shown an embodiment of the invention in which a vessel 4 containing the fluidised bed comprises a section of relatively small cross-sectional area 31, a section of relatively large cross-sectional area 32 and a transitional section 33 of frusto-conical shape. This arrangement causes a decrease in the linear velocity of gases above the upper surface of the fluidised bed 1 when the apparatus is in use so that any oversize particles of particulate material being heat treated, or any particles of inert refractory material which have been carried out of the fluidised bed, are returned to the fluidised bed.

Referring next to Figure 4, there is shown an embodiment of the invention in which a vessel 4 contains two fluidised beds 1a and 1b which are supported on perforated distribution plates 2a and 2b respectively. Fuel for combustion in the vessel, e.g. fuel oil, can be injected into either or both of the fluidised beds through conduits 7a and 7b. The particulate material which is to be heat treated is injected, preferably in suspension in air, into either or both of the fluidised beds through conduits 8a and 8b. Discharge conduits 15a and 15b permit discharge of the inert refractory material from the vessel.

The invention is further illustrated by the following Examples.

EXAMPLE 1.

A ground kaolinitic clay mineral, from which particles having an equivalent spherical diameter greater than 5 microns had been removed by gravitational and centrifugal separation and which comprised 80% by weight of particles smaller than 2 microns equivalent spherical diameter, was fed pneumatically into the apparatus described above with reference to Figure 1 of the accompanying drawings. The apparatus contained a fluidised bed having a diameter of $10\frac{1}{2}$ inches and containing approximately 30 lbs. of MOLOCHITE which was retained in, and which constituted the major part of, the fluidised bed throughout the process. The MOLOCHITE had a particle size distribution such that substantially all the particles passed through a $\frac{1}{8}$ of an inch mesh sieve and were retained on a No. 8 mesh British Standard sieve. The pressure drop across the fluidised bed was 10 inches water gauge. The initial inlet temperature of combustion gases to the fluidised bed was 1480° C., and the operational temperature of the fluidised bed was 78 cubic feet per minute, and the feed rate of ground kaolinitic clay mineral was 61 lbs/hr. Under these conditions, the calcined material had the properties noted in Table I below, which are compared with the initial properties of the kaolinitic clay mineral.

TABLE I

Property	Before Treatment	After Treatment
Valley abrasion value	30.	101
Specific gravity	2.64	2.14
Loss on ignition (weight %)	13.1	0.2
Percent by weight smaller than 2 microns	80	22
Brightness*	89.5/93.0	89.1/91.9

*Measured as % reflectance to light of 458 mµ/574 mµ wavelength on an Elrepho brightness meter.

EXAMPLE 2.

A kaolinitic clay mineral similar to that used in Example 1 above was treated as described in Example 1, except that the initial inlet temperature of combustion gases to the fluidised bed was 1180° C., the bed temperature was 900° C., the air flow through the fluidised bed was 90 cubic feet per minute, and the feed rate of kaolinitic clay mineral was 55 lbs/hr. The results obtained are set out in Table II below.

TABLE II

Property	Before Treatment	After Treatment
Valley abrasion value	30	101
Specific gravity	2.64	2.30
Loss on ignition (weight %)	13.1	0.5
Percent by weight smaller than 2 microns	80	19
Brightness*	89.5/93.0	86.7/90.7

*Measured as % reflectance to light of 458 mμ/574 mμ wavelength on an Elrepho brightness meter.

EXAMPLE 3.

A kaolinitic clay mineral similar to that employed in Examples 1 and 2 above was treated as described in Example 1, except that the inlet temperature of combustion gases to the fluidised bed was 920° C., the bed operating temperature was 700° C., the air flow through the fluidised bed was 108 cubic feet per minute, and the feed rate of kaolinitic clay mineral was 65 lbs/hr. The results obtained are set out in Table III below.

TABLE III

Property	Before Treatment	After Treatment
Valley abrasion value	30	109
Specific gravity	2.64	2.46
Loss on ignition (weight %)	13.1	2.0
Percent by weight of particles smaller than 2 microns	80	21
Brightness*	89.5/93.0	84.3/88.6

*Measured as % reflectance to light of 458 mμ.574 mμ wavelength on an Elrepho brightness meter.

EXAMPLE 4.

A kaolin clay comprising 80% by weight of particles smaller than 2 microns equivalent spherical diameter was treated as described in Example 1 above, except that the initial inlet temperature of combustion gases to the fluidised bed was 1250° C., the bed temperature was 1000° C., the air flow through the fluidised bed was 85 cubic feet per minute, and the feed rate of kaolin was 55 lb/hour. The calcined product was air classified and the fine product of the classification process was found to have a particle size distribution such that 34% by weight consisted of par-

ticles smaller than 2 microns equivalent spherical diameter, 8% by weight of particles larger than 10 microns equivalent spherical diameter and 0.02% by weight of particles larger than 53 microns. The specific gravity was 2.15, the loss on ignition 0.647% by weight, the percentage reflectance to light of 458 nm. wavelength was 86.5 and to light of 574 nm. wavelength was 89.8. This mineral is referred to hereinafter as " Filler A ".

A second mineral filler, hereinafter referred to as "Filler B ", was a kaolin clay, having the same particle size distribution, brightness and rheological properties as that used to prepare Filler A, which had been calcined in a conventional multiple hearth furnace at a maximum temperature of 950° C. for a total time of 4 hours and the final product has a particle size distribution such that 10% by weight consisted of particles having an equivalent spherical diameter larger than 10 microns and 50% by weight consisted of particles having an equivalent spherical diameter smaller than 2 microns. The specific gravity was 2.50 and the percentage reflectance to light of 458 nm wavelength was 88.0.

Each of the two fillers A and B were incorporated into butyl rubber compounds A and B according to the recipes given in Table IV below: —

TABLE IV

Component	Parts by weight	
	Compound A	Compound B
Butyl rubber (0.65 mole % unsaturation)	100	100
Process oil	5	5
Paraffin wax	5	5
Zinc oxide	5	5
Sulphur	1	1
p-quinone dioxime dibenzoate	6	6
Red lead	9	9
Filler A	108	—
Filler B	—	130

Batches of each of the two compounds were cured in a steam heated press at 160° C. for 40 minutes and the cured sheets were subjected to tests for modulus at 300% elongation, tensile strength, % elongation at break, hardness (in International Rubber Hardness Degrees) and tear strength. The tests were all carried out in accordance with B.S. 903 and the results are given in Table V below.

TABLE V

Property	Compound A	Compound B
Modulus at 300% elongation lbf/in² (Kgf/cm²)	375 (26.4)	390 (27.4
Tensile strength lbs/in² (Kgf/cm²)	540 (38.0)	515 (36.2)
Elongation at break %	555	515
Hardness IRHD (see BSS 903:Part A7 = 1957)	60	58
Tear strength lbf (Kgf)	11.1 (5.03)	11.1 (5.03)

These results show that there is no significant difference in mechanical properties between Compound A and Compound B. However, 5 Filler A has a lower specific gravity than Filler B and therefore a smaller weight is required to fill a given volume. Rubber fillers are generally bought by weight and therefore a saving in cost may be effected. In addition 10 Filler A has a slightly different refractive index from that of the rubber matrix and therefore has some pigmenting properties. Filler B has a refractive index which is substantially the same as that of the rubber matrix. 15

The cured compounds were also subjected to tests for electrical insulation properties after immersion in water at 50° C. in accordance with B.S. 2899 part 3. The results are shown in Table VI below. 20

TABLE VI

	Compound A	Compound B
% increase in capacitance after immersion at 50°C between		
1st & 14th days	7.1	5.3
7th & 14th days	5.3	2.5
Permittivity after 14 days immersion at 50°C.	4.3	4.1
Power factor after 14 days immersion at 50°C.	0.011	0.011

There is no significant difference between the results for Compound A and Compound B.

There is no significant difference between the results for Compound A and Compound B.

EXAMPLE 5.

25   A kaolin clay comprising 80% by weight of particles smaller than 2 microns equivalent spherical diameter was treated as described in Example 1 above, except that the initial inlet temperature of combustion gases to the fluidised bed was 1250° C., the bed 30 temperature was 1000° C., the air flow through the fluidised bed was 85 cubic feet per minute, and the feed rate of kaolin was 55 lb/hr.

The final product had a specific gravity of 2.2, a loss on ignition of 0.025% by weight, 35 a percentage reflectance to light of 458 nm. wavelength of 87.2 and to light of 574 nm. wavelength of 91.9 and a particle size dis-

tribution such that 23% by weight consisted of particles smaller than 2 microns equivalent spherical diameter, 12% by weight consisted of particles larger than 10 microns equivalent spherical diameter and 0.03% by weight consisted of particles larger than 53 microns. This material is hereinafter referred to as "Extender A".

A second mineral material, hereinafter referred to as "Extender B", was a kaolin clay, having the same particle size distribution, brightness and rheological properties as that used to prepare Extender A, which had been flash calcined in a conventional manner by passing the clay in powder form rapidly through an oil-fired combustion chamber at 850° C. The material had a specific gravity of 2.0 and a particle size distribution such that 35% by weight consisted of particles smaller than 2 microns equivalent spherical diameter.

Two exterior grade emulsion paints were made up according to the following recipe.

Paint A contained Extender A and Paint B contained Extender B.

	% by weight
Rutile titanium dioxide	20.6
Extender A or B	12.6
2% solution of hydroxy ethyl cellulose	17.4
5% dispersant solution	3.9
Butyl Carbitol acetate ("CARBITOL" is a Registered Trade Mark)	1.0
Water	10.9
Vinyl acetate/vinyl Versatic acid ester copolymer at 50% by weight solids ("VERSATIC" is a Registered Trade Mark)	33.5

Small quantities of anti-freeze, rust inhibitor, fungicide and defoamer.

The pigment volume concentration was 40%. The two paints were subjected to tests for film brightness, dispersion and opacity (contrast ratio) and the results are given in Table VII below.

TABLE VII

Property	Paint A	Paint B
Film brightness[1] %(MgO = 100%)	89.3	88.2
Dispersion[2] μm	25	35
Opacity[3] (contrast ratio)	93.0	92.3

Notes:

1. The film brightness test comprised applying a film of the paint at a constant thickness of 0.0032 inch (0.081 mm) to a sheet of "MELINEX" (Registered Trade Mark) plastics material and cutting five discs from the coated sheet. The five discs were stacked and five measurements of the brightness of the top disc of the stack were made using an ELREPHO brightness meter with light of 457 nm. wavelength. Between each measurement the bottom disc of the stack was transferred to the top, and the average of the five measurements was calculated.

2. The dispersion test was performed by drawing down each paint on a Hegman gauge. Both paints had previously been mixed in the same laboratory SILVERSON (Registered Trade Mark) shrouded-impeller mixer for a standard time of 25 minutes.

3. The opacity test comprised applying a film of the paint at a constant thickness of 0.0032 inches (0.081 mm) using a draw bar over Morest charts No. 102 which have equal black and white portions. The percentage reflectance (MgO=100%) to light of 540 nm wavelengths of the film over the black and white portions were measured with an ELREPHO brightness meter. The contrast ratio was calculated by the formula:

$$\text{contrast ratio} = \frac{\%\ \text{reflectance over black portion}}{\%\ \text{reflectance over white portion}} \times 100$$

Additional qualitative tests were performed to compare the brushability, flow, visual opacity, visual brightness and can stability of the two paints. Paint A was observed to have superior brushability, flow, visual opacity and visual brightness properties to Paint B, but the two paints had similar good can stability.

WHAT WE CLAIM IS:—

1. A process for heat treating a particulate material which consists predominantly of particles smaller than 50 microns equivalent spherical diameter, which process comprises passing the particulate material through a heated fluidised bed comprising particles of an inert refractory material which range in diameter from 0.5 to 5.0 mm, wherein (i) the fluidised bed contains throughout the period in which the particulate material is passing therethrough a major proportion by weight of said inert refractory material; (ii) the size, shape and density of the particles of inert refractory material and the velocity of the fluidising gas are such that the particles of inert refractory material are retained in the fluidised bed while the particulate material is carried through the fluidised bed; and (iii) the particulate material is retained within the fluidised bed for a time sufficient to effect the desired heat treatment thereof.

2. A process according to Claim 1, wherein the particle size distribution of the inert refractory material is such that the coarsest particles of the inert refractory material are not more than four times larger than the finest particles of the inert refractory material.

3. A process according to Claim 1 or 2, wherein the shape and the average particle size of the inert refractory material are such that the linear gas velocity necessary to fluidise the inert refractory material is at least five times that necessary to convey out of the fluidised bed the particulate material which is to be heat treated.

4. A process according to Claim 1, 2 or 3, wherein the particulate material to be treated is fed pneumatically into the fluidised bed.

5. A process according to Claim 1, 2, 3 or 4, wherein the particulate material to be treated is fed into the fluidised bed via a conduit passing longitudinally through the top of the reactor which contains the fluidised bed.

6. A process according to Claim 1, 2, 3, 4 or 5, wherein the inert refractory material comprises a sand, silica or a ceramic material.

7. A process according to Claim 6, wherein the inert refractory material comprises a calcined kaolin clay.

8. A process according to any one of the preceding claims, wherein the particulate material to be treated comprises a silicate of aluminium or of an alkaline earth metal.

9. A process according to Claim 8, wherein the particulate material to be treated comprises a kaolinitic clay.

10. A process according to any one of the preceding claims, wherein the fluidised bed is operated at a temperature of from 600° C. to 1200° C.

11. A process according to any one of the preceding claims, wherein the fluidised bed is heated by hot combustion gases.

12. A process according to any one of Claims 1 to 10, wherein the fluidised bed is heated by burning a fuel therein.

13. A process according to any one of the preceding claims, wherein the fluidised bed is initially heated by passing hot combustion gases therethrough until the temperature of the fluidised bed approaches the desired working temperature and thereafter the supply of hot combustion gases is stopped and a fuel is injected into the fluidised bed.

14. A process according to any one of the preceding claims, wherein the average residence time of the particulate material being treated in the fluidised bed is not more than one second.

15. A process according to any one of the preceding claims, wherein there are employed two fluidised beds in series.

16. A process according to any one of the preceding claims, wherein the particulate material is separated from the fluidising gas after passing through the (or each) fluidised bed by means of one or more cyclones.

17. A process according to Claim 16, wherein the ssupension of particulate material in the fluidising gas leaving the fluidised bed is cooled by the injection of a cold fluid into said suspension.

18. A process, according to claim 1, for heat treating a particulate material, substantially as hereinbefore described with reference to the accompanying drawings.

19. An apparatus for use in the process of Claim 1 substantially as hereinbefore described with reference to, and as illustrated in, the accompanying drawings.

20. A particulate material whenever treated by a proceess or in an apparatus as claimed in any one of the preceding claims.

21. A particulate material as claimed in Claim 20, wherein said particulate material is a silicate of aluminium or of an alkaline earth metal.

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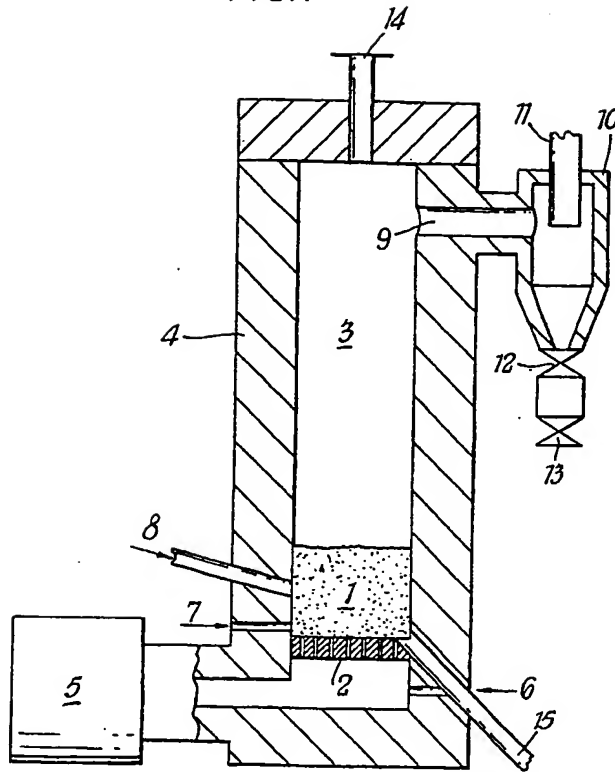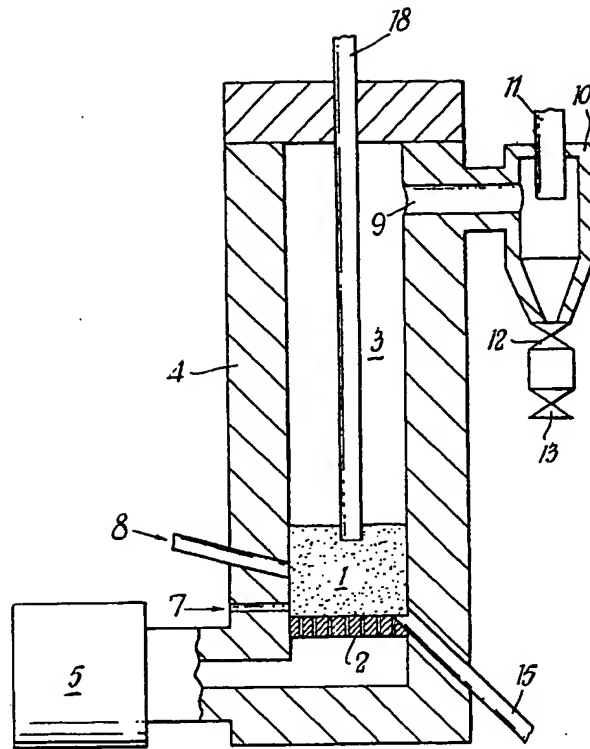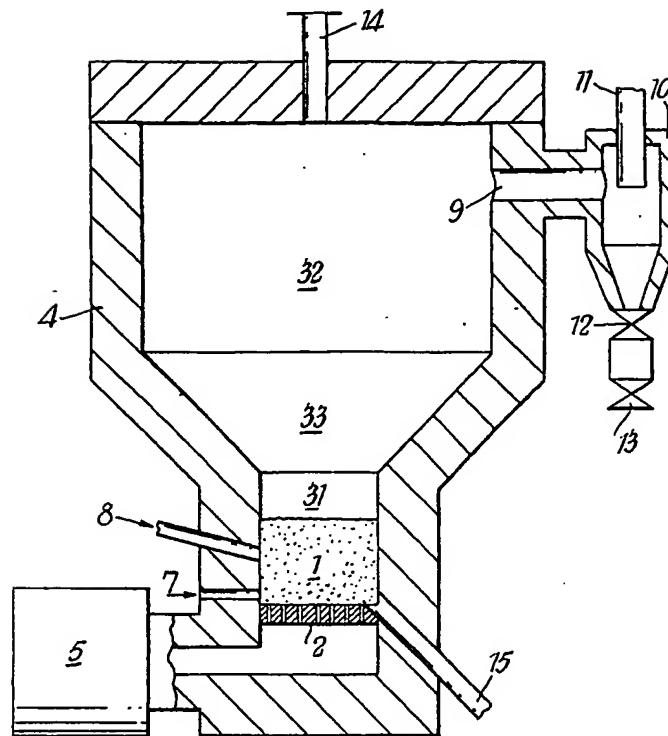London, W.C.2A 1AT.
Agents for the Applicants.

FIG.1

FIG.2

FIG.3

FIG.4